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Synthesis and Thermal Stability of Graphite Oxide-Like Materials

Ching-cheh Hung
Lewis Research Center
Cleveland, Ohio

Jean Corbin
Cleveland State University
Cleveland, Ohio

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SYNTHESIS AND THERMAL STABILITY OF GRAPHITE OXIDE-LIKE MATERIALS

Ching-cheh Hung
National Aeronautics and Space Administration
Lewis Research Center
Cleveland, Ohio 44135

and

Jean Corbin
Department of Chemistry
Cleveland State University
Cleveland, Ohio 44115

SUMMARY

Graphite oxide is typically made in a process where crystalline graphite was mixed with H_2SO_4, $NaNO_3$ and $KMnO_4$ for overnight reaction, then mixed with water for further reaction, and finally rinsed with methanol. In this report, crystalline graphite was substituted by submicron graphite powder, pitch-based graphitized carbon fibers, and activated carbon as the carbon reactants in this process. The reactions produced graphite oxide-like material. They were amorphous, but contained oxygen in the concentration range of the traditional graphite oxide. The weight, chemical composition, and structures of these material were characterized before and after they were exposed to high temperature nitrogen. The data thus obtained were then used to calculate the carbon and oxygen loss during heating. They began to lose both water and carbon at a temperature below 200 °C. During such decomposition, the lower the degree of graphitization, the higher the contribution of carbon loss to total mass loss. Also, slower heating when the temperature was lower than 150 °C resulted in lower carbon loss and higher oxygen loss. For example, slowly heating the sample (<1 °C/min.) made from activated carbon to 150 °C produced nearly no carbon loss, but 53 percent oxygen loss. Complete oxygen removal from the sample, however, has never been observed in this study, in which some samples were heated to 1000 °C. The same method was used to treat 0.254 mm thick graphite sheet. Instead of a graphite oxide-like material, an intercalation compound was produced. The graphite oxide-like materials obtained using activated carbon, crystalline graphite and submicron graphite powder as precursors all reacted with $AlCl_3$. The highest Al:C atomic ratio in the products was estimated to be 1:1.7. This implies the possibility of applications of this process in the area of batteries, catalysts, and sensors.

1. INTRODUCTION

High surface area ceramic products, alone or in carbon, could potentially be useful in the areas of batteries, catalysts, and sensors. One of such high surface area ceramic product (Al_2O_3) was produced in a recent study according to the following reactions (ref. 1):

$$\text{Graphite} \longrightarrow \text{Graphite oxide} \tag{1}$$

$$\text{Graphite oxide} + AlCl_3 \xrightarrow[\text{nitrogen}]{100\text{–}180\ ^\circ C} [C(Al, Cl, O) + \text{unreacted } AlCl_3]_{\text{mixture}} \tag{2}$$

$$[C(Al, Cl, O) + \text{unreacted } AlCl_3]_{\text{mixture}} \xrightarrow[\text{nitrogen}]{300\text{–}400\ ^\circ C} [AlCl_3]_{\text{vapor}} + C(Al, Cl, O) \quad \text{atomic Al/C} = 1{:}4 \tag{3}$$

$$C(Al, Cl, O) \xrightarrow[\text{air}]{300\text{–}500\ ^\circ C} Al_2O_3 \quad \text{surface area} = 80\ m^2/gm \tag{4}$$

where graphite oxide in reaction (1) was made from crystalline graphite powder according to the method developed by Hummers and Offeman (ref. 2), and C(Al, Cl, O) in reaction (2) was carbon containing aluminum, chlorine, and oxygen. It is noted that in C(Al, Cl, O), the Al:C atomic ratio was as high as 1:4. It is also noted that the final product (Al_2O_3) had a high surface area of 80 m^2/gm (ref. 1).

The geometric shape of the Al_2O_3 product in reaction (4) follows that of the carbon. For example, when the graphite in reaction (1) was graphitized carbon fibers (P-100), the final Al_2O_3 product in reaction (4) was fibrous.

It is hoped that the above-described reactions (1) to (4) can be generalized so that the products in reaction (4) is not limited in Al_2O_3 powder or fibers. For this purpose, it was suggested that nongraphitized, less expensive carbon materials could be used to replace crystalline graphite in reaction (1), and other metal halides could be used to replace $AlCl_3$ in reaction (2). Using metal halides other than $AlCl_3$ would result in ceramic products other than Al_2O_3. Using carbon materials other than crystalline graphite powder may have the advantage of reducing the cost and obtaining ceramic products of desirable geometric shape. For example, if commercially available activated carbon with surface area of 1000 m^2/g could be used as the precursor of these reactions, the cost of carbon could be reduced, and the surface area of the final ceramic product could be greatly increased.

The possibility of using metal chloride other than $AlCl_3$ for the above reactions was demonstrated previously (ref. 1). However, the possibility of using nongraphitized carbon materials for the process described above is not known. In fact, graphite has been the only carbon material reported as the reactant for graphite oxide synthesis since its discovery in 1860 (ref. 3).

The purpose of this report is to describe the possibility to make products similar to graphite oxide by using nongraphitized carbon materials to substitute crystalline graphite in the graphite oxide synthesis process, and to characterized the product thus obtained. This is to set the foundation for future work of making high surface area ceramic materials using nongraphitized carbon as a template. In this research, today's commercially available nongraphite carbon materials were used to substitute crystalline graphite in the graphite oxide synthesis process. The products were compared to the graphite oxide made from crystalline graphite. Their structure, chemical composition, thermal stability and reactivity to $AlCl_3$ were examined.

2. EXPERIMENTAL

2.1. Synthesis Reactions

The method developed by Hummers and Offeman (ref. 2) was used to synthesize graphite oxide-like materials. In this method, the carbon materials were mixed with H_2SO_4, $NaNO_3$ and $KMnO_4$ for overnight reaction, then mixed with 100 °C water for further reaction, and finally rinsed with methanol.

2.2. Carbon Materials

In this research five different kinds of carbon materials were used to react with these chemicals. They were, in the order of decreasing x-ray diffraction (XRD) peak height, crystalline graphite powder (300 mesh, 99 percent), graphite powder (<1 μm, 99.9995 percent), graphite sheet (0.254 mm thick, 99.9 percent), graphitzed carbon fiber (Amoco VCB-45, 10 μm diam., 99 percent), and activated carbon (1100 m^2/gm, containing small amounts of Al and Si).

2.3. Heating experiments

All graphite oxide-like materials thus obtained were simultaneously heated in nitrogen in the same heating apparatus. Three such runs were made. The temperature was maintained at 100, 150, and 200 °C, respectively, for 20 to 24 hr. The heating rates were such that the samples heated to 100 and 150 °C reached the final temperature in 16 min, and the sample heated to 200 °C reached 180 °C in 90 min, and then reached 200 °C in another 90 min. This set of experiments resulted in graphite oxide-like materials obtained from different precursors and subsequently experienced identical temperature histograms.

The graphite oxide-like samples made from submicron graphite powder, VCB-45 fibers, and activated carbon were also heated to the higher final temperature of 270, 385, and 650 °C. They were at first heated to 200 °C by the above-described temperature histogram before heated to the final temperature and kept at the final temperature for 20 to 24 hr.

The sample made from crystalline graphite was not used in this part of the experiment involving heating to a temperature higher than 200 °C because in several such attempts, the sample exploded (i.e., disintegrated into fine powder and some gaseous products in a fraction of a second) at about 200 °C. In one such attempt, it was heated with those made from submicron graphite powder and activated carbon. The heating rate was about 1 °C/min. The sample made from crystalline graphite exploded when its environment reached 210 °C. The other two samples did not explode and were kept at 250 °C for 5 hr before being heated to 1000 °C over a 1-hr period.

Heating the graphite oxide sample made from crystalline graphite to 1000 °C without explosion was finally accomplished by heating a small sample (3 mg) to 210 °C in 90 min, holding the temperature at 210, 260, 360 and 460 °C for 0.5, 2, 1 and 5.5 hr, respectively, before heating to 1000 °C over a 30 min period. Since the sample was small, the chemical composition of this product was not able to be determined.

To demonstrate the effects of heating rate, the products obtained from heating graphite oxide-like samples to the same final temperature, but at different heating rates, were compared. The amount of carbon loss during heating was used as the criterion in stability comparison. In this research, such comparison was made between four samples obtained from heating graphite oxide-like material made from activated carbon at two different heating rates (<1 °C/min as slow heating, >1 °C/min as fast heating) to two different final temperatures (150 and 650 °C). Among all carbon materials used in this research, the activated carbon precursor was selected for this heating rate study because, as will be described later in this report, its graphite oxide-like product was found to have highest (and therefore most accurately measured) carbon loss during heating.

2.4. Characterization

A "Leco" process, named after the company who developed it, was used to measure the bulk carbon and sulfur mass percent. In the process, carbon and sulfur in the samples were converted into CO_2 and SO_2, respectively, which were then quantitatively measured using their properties of IR absorption [ref. 4].

The bulk metal contents were measured by inductively coupled plasma mass spectrometry (ICPMS). The surface compositions were analyzed using x-ray photoelectron spectroscopy (XPS). The structures were studied using x-ray diffraction (XRD) data. The microscopic views were obtained from scanning electron microscopy (SEM). The identification of the chemical elements in the samples, as well as the semiquantitative chemical analyses, were conducted using their energy dispersive spectra (EDS).

The sample mass before and after heating was measured. The carbon mass loss during heating was estimated as follows:

Given the reaction:

$$\text{Graphite oxide-like material} \xrightarrow[\text{Heat in Nitrogen}]{} \text{Decomposed solid Product + gas} \qquad (5)$$

Let the graphite oxide-like (reactant) have mass M_r, carbon content X_r, and therefore carbon mass M_rX_r. Similarly, the decomposed solid product have mass M_p, carbon content X_p, and therefore carbon mass MpX_p. Therefore,

$$\text{Carbon mass loss} = M_rX_r - M_pX_p, \qquad (6)$$

$$\text{Carbon mass loss as a fraction of reactant carbon} = (M_rX_r - M_pX_p)/M_rX_r = [X_r - (M_p/M_r)\,X_p]/X_r, \qquad (7)$$

and

$$\text{Carbon mass loss as a fraction of total mass loss} = (M_rX_r - M_pX_p)/(M_r - M_p) = [X_r - (M_p/M_r)X_p]/[1 - (M_p/M_r)], \qquad (8)$$

where X_r and X_p, the percent carbon weight in the reactant and the solid product, respectively, can be obtained by chemical analysis, and

$$[1 - (M_p/M_r)] = (M_r - M_p)/M_r = \text{percent mass loss during reaction} \quad (9)$$

2.5. Reaction to $AlCl_3$

The products obtained from the precursors of crystalline graphite powder, submicron graphite powder, and activated carbon were exposed to $AlCl_3$ to test their ability to hold large quantities of aluminum. They are mixed with the commercially purchased $AlCl_3$ powder at a temperature histograms between 120 and 193 °C for 42 hr, and finally the sample was held at 225 °C for 0.2 hr to evaporate the unused $AlCl_3$.

The graphite oxide-like material made from activated carbon was also exposed to saturated $AlCl_3$ vapor. This sample and a sample of its activated carbon precursor were placed side by side in a tubular reactor during the reaction. The temperature histogram was 125 to 175 °C for 46 hr, then 200 °C for 39 hr, and finally 200 to 300 °C for one hour. Higher temperature and extra time were applied to remove unreacted $AlCl_3$ in a relatively large sample of activated carbon.

3. RESULTS AND DISCUSSION

3.1. SEM observation

All carbon materials reacted during the graphite oxide synthesis process described above. Figure 1 shows the micrographs of these products obtained from SEM. The samples were not treated with conductive coating. The graphite oxide sample obtained from crystalline graphite move during SEM examination. This was the result of the electron beam on the insulative product. For this sample, the micrographs in figure 1 were obtained from a non-typical small area where no such movement occurred.

3.2. Chemical composition

Chemical analysis of the products of the graphite oxide-like material synthesis reactions is described in table I. Note that the column of carbon content gives the values of X_r described in equations (6) to (8). All samples contained large quantities of oxygen. In many cases the atomic oxygen to carbon ratio were higher than the theoretical maximum of 0.5. This is because of the presence of water in the samples.

The product made from 300 mesh crystalline graphite powder appears nonuniform. There were clay-like large and small aggregates which appeared wet and sand-like small particles which appeared dry. The nonuniformity of this sample made the composition analysis difficult. Since the sample composition data were used to calculate the carbon loss, the carbon loss data for the graphite oxide made from crystalline graphite is questionable. This will be further discussed in Section 3.4.2.

The product made from graphite film also was not uniform. All other graphite oxide-like products appeared uniform in terms of color and particle size.

3.3. Structure

The XRD data indicated that all of the carbon reactant had graphite peaks, but none of these products had graphite peaks. The sample made from graphite sheet had sharp peaks at 5.23 and 2.62 Å. Together with their high sulfur content (table I), it is believed to be a H_2SO_4–graphite intercalated compound with an identity period of 5.23 or 10.46 Å. This sample was not a graphite oxide-like compound and was not studied further.

The sample made from crystalline graphite was the only one that had the graphite oxide peak at about 8 Å. All other samples has no XRD peaks, suggesting amorphous structures. Since they did not have graphite oxide peaks but are compounds of carbon and oxygen, they are called graphite oxide-like materials.

3.4. Thermal stability

3.4.1. Disintegration.—"Explosion" is believed to be a result of fast temperature rise that causes and is caused by a highly exothermic carbon-oxygen reaction, therefore producing gases (CO or CO_2) quickly. Based on this hypothesis, the explosion can be avoided if the heat generated by the reaction can be either reduced or quickly removed. This can be done if the heating rate is slow and/or if the graphite oxide sample is small, has low oxygen-carbon ratio, has a small carbon crystallite or has a large surface area.

In this study, upon heating, the graphite oxide made from crystalline graphite was the only product that exploded. This is probably because it had large carbon crystallites and high oxygen-carbon ratio. No explosion was found for the graphite oxide-like materials produced in this study.

3.4.2. Total Mass Loss and Carbon Loss.—Table II describes the mass loss of the graphite oxide and graphite oxide-like materials during their thermal decomposition (i.e., the $[1 - (M_p/M_r)]$ values described in equations (9)). This table shows that significant quantities of masses were lost at a temperature of as low as 100 to 200 °C. It is believed that large fractions of these mass losses were due to water evaporation. This table also shows that, at a given heating temperature, graphite oxide-like material made from more graphitized carbon loses more percent mass. This, however, does not mean that graphite oxide-like material made from more graphitized carbon precursor is less thermally stable because their large quantities of mass loss were mostly water. This table, however, shows that graphite oxide-like material continues to thermally decompose even when the temperature is raised to as high as 1000 °C.

The thermal stability of these graphite oxide-like materials is best examined by estimating the carbon loss during heating. These values can be calculated from equations (7) and (8), where the carbon content (X_r) are described in table I, the mass loss data $[1 - (M_p/M_r)]$ at 200 °C are described in table II, and the carbon contents of the graphite oxide-like samples after the 200 °C heating (X_p) are described in table III. Using these data and equations (7) and (8), the carbon loss during heating at 200 °C were estimated and summarized in table IV. It is observed that carbon loss began to occur at a temperature below 200 °C for the products made from nongraphite carbon, but above 200 °C for those made from crystalline graphite. At 200 °C, the lower the degree of graphitization, the higher the contribution of carbon loss to total mass loss.

In table IV, the negative value of carbon loss for the graphite oxide made from crystalline graphite appears to be a result of inaccurate carbon content described in table I. As described earlier, this graphite oxide was not uniform. Therefore, the sample used for chemical analysis may not be accurate. However, knowing that the graphite oxide made from graphite loss little carbon at a temperature below 200 °C (ref. 5), regardless the accuracy of this data point, the trend described in the last paragraph is believed to be accurate.

3.4.3. Effects of heating rate on the thermal stability.—Table V summarizes the results of slowly heating graphite oxide-like material made from activated carbon to 150 and 650 °C. The heating rate was less than 1 °C/min when the samples were cooler than 150 °C. Again the carbon loss data described in table V were obtained using equations (6) to (9). The oxygen loss data were obtained using the same method. For demonstrating the effects of heating rate when the samples were cooler than 150 °C, the data obtained from fast heating the sample to 150, 200, and 650 °C are also included in this table. It can be seen from this table that fast heating produced higher carbon loss as well as total weight loss. In other words, the graphite oxide-like materials were less thermally stable if the heating rate was higher. In a specific example, the carbon loss for slow heating to 150 °C was only 2.5 percent, while the oxygen loss was 53 percent. In another example, the carbon loss for slow heating (<1 °C/min) to 650 °C was about 15 percent, the same for fast heating (2 °C/min) to 200 °C. On the other hand, the oxygen loss for slow heating (<1 °C/min) to 650 °C was about 74 percent, much more than the value of 43 percent for fast heating (2 °C/min) to 200 °C. However, it is noted that after 650 °C heating, decomposition of this graphite oxide-like material was not complete. There was still significant quantity of oxygen left in the carbonaceous sample.

3.5. Reaction With $AlCl_3$

The graphite oxide-like material made from activated carbon precursor was exposed to $AlCl_3$ vapor and resulted in 49 percent weight gain. The activated carbon precursor that was exposed to $AlCl_3$ at the same time had 36 percent weight gain. The ability of this carbon material to pick up $AlCl_3$ increased after this oxidation treatment.

After the reaction with $AlCl_3$, the chemical composition in the samples was measured using Leco and ICPMS. Three samples were used in this set of experiments: Samples made from activated carbon precursor and exposed to

$AlCl_3$ vapor, samples made from crystalline graphite precursors and mixed with $AlCl_3$ powder, and sample made from submicron graphite powder and mixed with $AlCl_3$. The Al:C atomic ratios were approximately 1:7, 1:4, and 1:1.7, respectively. The results are summarized in table VI. Similar compositions were found from XPS analysis on the sample surfaces and were also summarized in table VI. The atomic Al:C ratio for the product made from the submicron graphite was found to be as high as 2:5 on the surface. This value is lower than the 1:1.7 value in the bulk. Therefore, the measured high aluminum concentration is not a result of a coating on the surface.

4. CONCLUSIONS

Graphite oxide-like materials were made using nongraphitized carbon materials to substitute crystalline graphite in the graphite oxide synthesis process. Such commercially purchased carbon materials included submicron graphite powder, pitch-based graphitized carbon fibers, and activated carbon. These graphite oxide like materials were amorphous, but contained oxygen in the concentration range of the traditional graphite oxide made using a process developed by Hummers and Offeman (ref. 2).

These samples began to lose both water and carbon at a temperature below 200 °C. During such decomposition, the lower the degree of graphitization, the higher the contribution of carbon loss to total mass loss. These graphite oxide-like material did not disintegrate into Å-sized fine powders at about 200 °C, as the graphite oxide made from crystalline graphite did.

Complete oxygen loss was not observed in this study, in which some samples were heated to 1000 °C.

Fast heating when the temperature was lower than 150 °C resulted in high carbon loss. If the heating rate was kept lower than 1 °C/min and the final temperature was 150 °C for 14 hr, the carbon loss was less than 2.5 percent for the graphite oxide-like sample made from activated carbon. This product had a chemical composition of $CO_{0.2}$.

The graphite oxide-like material obtained using activated carbon as the precursor reacted with $AlCl_3$, but to a lesser extent than that using crystalline graphite as the precursor. However, those made from submicron graphite powder reacted with $AlCl_3$ the most, resulting in a product having an Al:C atomic ratio as high as 1:1.7.

A specific example was the graphite oxide-like material made from activated carbon having surface area of 1100 m^2/gm. It reacted with $AlCl_3$ and resulted in a product having Al:C ratio of 1 to 7. The presence of such product paves the way for future works using relatively inexpensive, commercially purchased activated carbon materials as precursor to produce ceramics having surface area similar to that of activated carbon. Such high surface area ceramic products, alone or in carbon, could potentially be useful in the areas of batteries, catalysts, and sensors.

5. ACKNOWLEDGMENT

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TABLE I.—CHEMICAL ANALYSIS (WT %) OF THE PRODUCTS OF GRAPHITE OXIDE SYNTHESIS REACTIONS

Precursors	C	S	K	Mn	O[a] (balance)	atomic O/C ratio
Crystalline graphite	34.3	1.45	1.56	0.07	62.6	1.36
Submicron graphite	36.1	2.68	1.63	8.85	50.7	1.05
Graphite sheet	13.8	13.8	1.79	4.61	66.0	3.59
Carbon fiber	58.9	3.09	1.25	1.81	35.0	0.44
Activated carbon	61.1	1.96	0.46	0.04	36.4	0.45

[a]Includes water.

TABLE II.—MASS LOSS (% OF THE REACTANT MASS) DURING 100, 150, AND 200 °C DECOMPOSITION OF GRAPHITE OXIDE AND GRAPHITE OXIDE-LIKE MATERIALS

Precursors	atomic O/C ratio before heating	100 °C[a]	150 °C[a]	200 °C	270	385	650	1000
Crystalline graphite	1.36	24.4	46.7	41.5	——	——	——	61.8[b]
Submicron graphite	1.05	22.6	29.2	30.9	33.1	36.3	45.6	54.0
Graphite sheet	3.59	41.9	46.1	48.1	——	——	——	——
Carbon fiber	0.44	21.6	25.7	19.4	23.6	27.1	38.6	——
Activated carbon	0.45	26.4	29.7	25.2	29.1	33.1	44.6	56.9

[a]The low mass loss for some samples tested at 200 °C are believed to be the result of slow heating.
[b]This data point was obtained without explosion by heating a small sample (3 mg) to 210 °C in 90 min, kept at 210, 260, 360 and 460 °C for 0.5, 2, 1 and 5.5 hr, respectively before heating to 1000 °C in 30 min.

TABLE III.—CARBON CONTENT OF THE GRAPHITE OXIDE AND GRAPHITE OXIDE-LIKE SAMPLES AFTER 200 °C HEATING IN NITROGEN.

Precursors	Wt. % of Carbon after 200 °C heating in nitrogen	Atomic O/C ratio[a]
Crystalline graphite	63.8	0.42
Submicron graphite	49.9	[b]0.75
Carbon fibers	67.2	0.37
Activated carbon	70.3	0.32

[a]Numbers in this column were calculated assuming that carbon and oxygen were the only elements in the sample.
[b]Actual value is much lower than the calculated value of 0.75 because of the high Mn content described in table I was neglected.

TABLE IV.—CARBON LOSS DURING 200 °C DECOMPOSITION OF THE GRAPHITE OXIDE AND GRAPHITE OXIDE-LIKE MATERIALS

Precursors	% of Carbon mass before decomposition	% of Total mass loss during decomposition
Crystalline graphite	–8.8	–7.2
Submicron graphite	4.2	4.91
Carbon fibers	8.0	24.3
Activated carbon	14.1	34.1

TABLE V.—CHEMICAL COMPOSITION OF THE GRAPHITE OXIDE-LIKE MATERIAL OBTAINED USING ACTIVATED CARBON AS PRECURSOR

Temperature	Heating rate,[a] °C/min	Mass loss during heating, percent	Carbon content, percent	Oxygen content,[b] percent	Carbon remaining (percent of reactant carbon mass)	Oxygen remaining (percent of reactant oxygen mass)
No heating	----------	0	61.1	38.9	100	100
150 °C	<1	22.2	76.6	23.4	97.5	46.8
150 °C	9	29.7	—	—	—	—
200 °C	2	25.2	70.3	29.7	85.9	57.1
650 °C	<1	38.5	83.6	16.4	84.1	25.9
650 °C	2	44.6	—	—	—	—

[a] This is the heating rate when the temperature was in room temperature to 150 °C range.
[b] Assuming carbon and oxygen were the only elements in the samples.

TABLE VI.—CHEMICAL ANALYSIS (WT %) OF THE CARBONACEOUS PRODUCTS HAVING HIGH AL:C RATIO

Precursors		C	Al	S	K	Mn	Cl	O	Balance[a]
Crystalline graphite	Bulk	24.8	13.2	0.52	0.6	0.07	- - -	- - -	60.8
	Surface	27.4	19.8	0.5	0	0	19.8	32.5	- - -
Submicron graphite	Bulk	15.1	20.4	1.19	1.3	1.1	—	—	60.9
	Surface	21.4	21.5	1.7	0.8	0	19.0	35.6	- - -
Activated carbon	Bulk	38.3	12.5	0.38	0.6	0.04	- - -	- - -	48.2
	Surface	35.1	18.0	- - -	- - -	- - -	16.9	30.0	- - -

[a] The balance is mostly oxygen and chlorine.

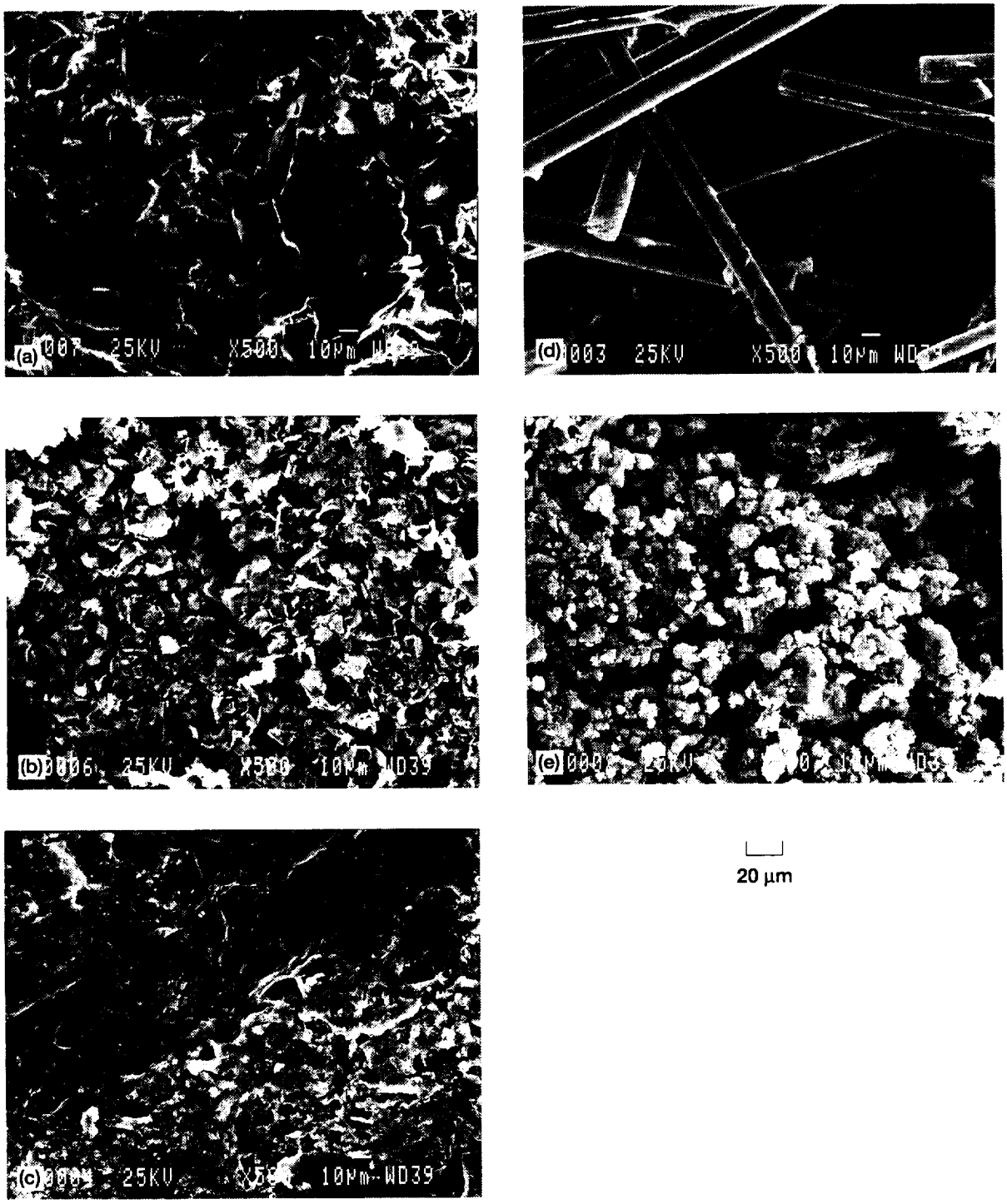

Figure 1.—SEM Micrographs of the products obtained from treating different precursors with the graphite oxide synthesis process. (a) Crystalline graphite. (b) Sub-micron graphite powder. (c) Graphite film. (d) VCB45 fibers. (e) Activated carbon.

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13. ABSTRACT (***Maximum 200 words***)

Graphite oxide is typically made in a process where crystalline graphite was mixed with H_2SO_4, $NaNO_3$ and $KMnO_4$ for overnight reaction, then mixed with water for further reaction, and finally rinsed with methanol. In this report, crystalline graphite was substituted by sub-micron graphite powder, pitch-based graphitized carbon fibers, and activated carbon as the carbon reactants in this process. The reactions produced graphite oxide-like material. They were amorphous, but contained oxygen in the concentration range of the traditional graphite oxide. The weight, chemical composition, and structures of these material were characterized before and after they were exposed to high temperature nitrogen. The data thus obtained were then used to calculate the carbon and oxygen loss during heating. They began to lose both water and carbon at a temperature below 200 °C. During such decomposition, the lower the degree of graphitization, the higher the contribution of carbon loss to total mass loss. Also, slower heating when the temperature was lower than 150 °C resulted in lower carbon loss and higher oxygen loss. For example, slowly heating the sample (<1 °C/min) made from activated carbon to 150 °C produced nearly no carbon loss, but 53% oxygen loss. Complete oxygen removal from the sample, however, has never been observed in this study, in which some samples were heated to 1000 °C. The same method was used to treat 0.254 mm thick graphite sheet. Instead of a graphite oxide-like material, an intercalation compound was produced. The graphite oxide-like materials obtained using activated carbon, crystalline graphite and sub-micron graphite powder as precursors all reacted with $AlCl_3$. The highest Al:C atomic ratio in the products was estimated to be 1:1.7. This implies the possibility of applications of this process in the area of batteries, catalysts, and sensors.

14. SUBJECT TERMS

Graphite oxide; Thermal stability; Ceramics in carbon

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